#2
11-30-00

PATENT

Docket No. CH9-1999-0004US1

jc542 U.S. PTO
09/614511
07/11/00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s)	: Wanda ANDREONI et al	Group Art:	not yet assigned
Serial No.	: not yet assigned	Examiner:	not yet assigned
Filed	: herewith		
For	: MATERIAL FOR USE IN A LIGHT-EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE		

CLAIM FOR PRIORITY UNDER 35 U.S.C. § 119

Honorable Commissioner of Patents and Trademarks
Washington, DC 20231

Sir:

A claim for priority is hereby made under the provisions of 35 U.S.C. § 119 for the above-identified U.S. patent application based upon European patent application number 99113398.4 filed July 12, 1999. A certified copy of this European patent application is filed herewith.

Respectfully submitted,

Stanley D. Ference III

Dated: June 11, 2000

Stanley D. Ference III
Reg. No. 33,879

FERENCE & ASSOCIATES
129 Oakhurst Road
Pittsburgh, Pennsylvania 15215
(412) 781-7386
(412) 781-8390-Facsimile

THIS PAGE BLANK (USPTO)

jc542 U.S. PTO
09/614511
07/11/00

Europäisches Patentamt	European Patent Office	Office européen des brevets

Bescheinigung / Certificate / Attestation

Die angehefteten Unterlagen stimmen mit der ursprünglich eingereichten Fassung der auf dem nächsten Blatt bezeichneten europäischen Patentanmeldung überein.

The attached documents are exact copies of the European patent application described on the following page, as originally filed.

Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr. / Patent application No. / Demande de brevet n°

99113398.4

Der Präsident des Europäischen Patentamts;
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets
p.o.

I.L.C. HATTEN-HECKMAN

DEN HAAG, DEN
THE HAGUE, 06/04/00
LA HAYE, LE

EPA/EPO/OEB Form 1014 - 02.91

THIS PAGE BLANK (USPTO)

Blatt 2 der Bescheinigung
Sheet 2 of the certificate
Page 2 de l'attestation

Anmeldung Nr.:
Application no.: 99113398.4
Demande n°:

Anmeldetag:
Date of filing: 12/07/99
Date de dépôt:

Anmelder:
Applicant(s):
Demandeur(s):
International Business Machines Corporation
Armonk, NY 10504
UNITED STATES OF AMERICA

Bezeichnung der Erfindung:
Title of the invention:
Titre de l'invention:
Material for use in a light emitting device and highly effecient electroluminescent device

In Anspruch genommene Prioriät(en) / Priority(ies) claimed / Priorité(s) revendiquée(s)

Staat: State: Pays:	Tag: Date: Date:	Aktenzeichen: File no. Numéro de dépôt:

Internationale Patentklassifikation:
International Patent classification:
Classification internationale des brevets:
/

Am Anmeldetag benannte Vertragstaaten:
Contracting states designated at date of filing: AT/BE/CH/CY/DE/DK/ES/FI/FR/GB/GR/IE/IT/LI/LU/MC/NL/PT/SE
Etats contractants désignés lors du depôt:

Bemerkungen:
Remarks:
Remarques:

THIS PAGE BLANK (USPTO)

SZ9-99-004

MATERIAL FOR USE IN A LIGHT EMITTING DEVICE AND HIGHLY EFFICIENT ELECTROLUMINESCENT DEVICE

EPO - Munich
52
12. Juli 1999

DESCRIPTION

The present invention relates to a material for use in a light emitting device. Specifically, the invention deals with an organic electroluminescent device. Still more specifically, the present invention relates to increasing the efficiency of organic light emitting devices (OLEDs).

BACKGROUND OF THE INVENTION

Electroluminescent devices based on organic thin layers are light-emitting devices similar to semiconductor-based light-emitting diodes, and today they are considered to be one of the flat panel displays of the next generation.

Such devices contain spaced electrodes separated by an electroluminescent medium that emits light in response to the application of an electrical potential difference across the electrodes.

In current preferred forms OLEDs are comprised of an anode, an organic hole injecting and transporting zone in contact with the anode, an electron injecting and transporting zone forming a junction with the organic hole injecting and transporting zone, and a cathode in contact with the electron injecting and transporting zone. When an èlectrical potential is placed across the electrodes, holes and electrons are injected into the organic zones from the anode and cathode, respectively. Light emission results from hole-electron recombination within the device. Such carrier recombination generates excited molecules, which eventually emit light or become thermally deactivated. Therefore, device efficiency is highly dependent on both carrier recombination efficiency and photoluminescence quantum yield of the emitting material. Increasing the efficiency of OLEDs is of primary importance to the acceptance of the new technology for displays of practical use.

Through intensive investigations and a series of recent inventions organic light emitting devices of improved characteristics, both in terms of fabrication feasibility and operating performance have been developed.

DESCRIPTION OF BACKGROUND ART

The commonly electroluminescent unit of a certain class of OLEDs (those based on small molecules) is a highly fluorescent aluminum complex, tris(8-quinolinolato)aluminum(III), (Alq_3), that emits in the green and fulfils a number of prerequisites for, e.g., the stability of the device and carrier transport but whose luminescense yield is relatively low. Previous attempts to improve it have been made by doping the Alq_3 layer with fluorescent dye molecules.

Kido et al., Appl. Phys. Lett., vol. 73, no. 19, pages 2721-2723, 9 November 1998, disclose efficient organic electroluminescent devices fabricated by using tris(4-methyl-8-quinolinolato)aluminum(III) ($Almq_3$) as an emitter layer. In addition to using this complex, a multilayer device structure, consisting of a hole-injecting layer, a hole transport layer, a dye-doped $Almq_3$ emitting layer, and an electron transport layer, was employed in order to reduce the driving voltage as well as to maximize carrier recombination efficiency. Kido et al. report a maximum luminance of over 140 000 cd/m^2 and an external quantum efficiency of 7.1%, which is said to be the highest efficiency ever reported for organic devices.

In US-A-5,150,006, there is disclosed an internal junction organic electroluminescent device comprised of, in sequence, an anode, an organic hole injecting and transporting zone, an organic electron injecting and transporting zone, and a cathode. The organic electron injecting and transporting zone is comprised of an electron injecting layer in contact with the cathode and, interposed between the electron injecting layer and the organic hole injecting and transporting zone, a blue emitting luminescent layer comprised of an aluminum chelate containing a phenolato ligand and two R^S-8-quinolinolato ligands, where R^S substituents are chosen to block the attachment of more than two substituted 8-quinolinolato ligands to the

aluminum atom. The presence of the phenolato ligand shifts the device emission to the blue region of the spectrum and increases emission efficiency. Device emission is shifted to even shorter blue wavelengths and increased operating stability can be realized by the incorporation of a pentacarboxyclic aromatic fluorescent dye.

US-A-5,456,988 discloses an electroluminescent (EL) device including an organic electron transport layer comprising Alq_3 substituted with Cl or Br in the 5-position. The object is to provide a useful EL device that has excellent durability and retains stable luminescence for a long period of time by using a compound other than 8-quinolinolato-aluminum complex as an emitting material. No values of luminance for the single halogen substitution are reported.

Therefore, there is still a need for the enhancement of the intrinsic luminescense of the organic molecular unit in Alq_3-based OLEDs.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide an organic material for use in a light emitting device showing an increased intrinsic luminescence of the organic molecular unit.

It is a further object of the invention to provide such a material that is based on Alq_3.

It is still a further object of the invention to provide a light emitting device with enhanced intrinsic luminescense.

These and other objects and advantages are achieved by the material disclosed in claim 1 and the device claimed in claim 8.

Preferred embodiments of the invention are described in the dependent claims.
To achieve the above mentioned objects, a new approach to the direct enhancement of the intrinsic luminescense of the organic molecular unit in Alq_3-based OLEDs is proposed. This is achieved by modifying the relevant electron states by means of the specific substitutes on the quinolate ring, namely by substituting the Alq_3 unit in positions 3 or 4 and 5 simultaneously using an electron-acceptor or p - delocalizing group in the 5-position and an electron-donor group in the 3- or 4- position.

This combined substitution amplifies the enhancement of the luminescence and reduces the induced shift of the ionization potential and electronic affinity values with respect to single substitutions. The latter is important to incorporate the new compounds in available device structures.

DESCRIPTION OF THE DRAWINGS

The invention is described in detail below with reference to the following schematic drawing. It is to be noted that the Figure is not drawn to scale.

FIG. 1 shows the formula of Alq_3 where the atoms on the quinolate ligand are labelled with the standard notation.

DESCRIPTION OF THE PREFERRED EMBODIMENT

The formula of Alq_3 is shown in Figure 1, where the atoms on the quinolate ligand are labelled with the standard notation.

From the theoretical work that was done, it has become clear that the relatively weak (compared with other fluorescent organic materials) luminescense of the Alq_3 molecule is associated with the different spatial localization of the electron states involved in the luminescence process which limits the corresponding transition probability. More in detail, the holes acceptor states (Highest Occupied Molecular Orbital (HOMO) set of states) are localized mainly on the phenoxyde side of the ligands whereas the electron acceptor states (Lowest Unoccupied Molecular Orbital (LUMO) set of states) are localized mainly on the pyridyl side of the ligands; this limits by fact the transition probability for HOMO - LUMO electronic transitions (the relevant ones for luminescence properties).

Thus, one can increase the intrinsic luminescense yield by modifying the relevant electron states by means of specific chemical substitutions on the quinolate rings, that improve the spatial overlap between the HOMO and LUMO set of states and therefore indirectly the transition probability and hence the intrinsic luminescence yield.

Proposed are substitutions using an electron-donor group (R^{py}) in the 3- or 4- position and, at the same time, an electron-acceptor or p- delocalizing group (R^{ph}) in the 5-position.

The substituents R^{py} can be selected from the groups -CR'R''R''', $-NR_2$, and -O-R, wherein R, R', R''= (H, Alkyl), R'''= (Alkyl) and generally from any group that is able "to push" electrons onto the ligands.

The substituents R^{ph} can be selected from the groups $-CX_3$, wherein X=F, Cl, Br, $-CX_2-CX_3$, $-SO_3-R$, $-CR=CR_2$, $-CX=CX_2$, -COOR, $-SO_3M$, and -COOM, wherein M = metal ion, R = alkyl and X = F, Cl, Br, and generally from any group that is able "to (strongly) suck"

electrons from the ligand or enhance the p-conjugation (delocalization) on the phenoxide-side of the ligand.

An electroluminescent device using the proposed Alq_3 derivatives consists of a hole injection electrode, an electron injection electrode and at least one organic emitting layer incorporating at least one of the proposed Alq_3 derivatives.

Moreover the electroluminescent device may contain additional hole-transport layers between the hole injection layer and the organic emitting layers and/or additional electron transport layers between the electron injection electrode and the organic light emitting layers.

Since the organic light emitting layer consists of Alq_3 derivatives having a larger intrinsic luminescence yield (the calculated enhancement factor goes up to 4) the device will have a larger quantum efficiency than devices made by unsubstituted and undoped Alq_3.

Moreover since this is obtained by directly modifying the Alq_3 molecule and without adding any highly fluorescent dopants (contrary to the work known from prior art), the good stability and carrier transport properties of Alq_3 layers are preserved, and no additional energy transfer step from Alq_3 to the dopant molecules is needed to have high luminescence yield.

SZ 9-99-004

EPO - Munich
52
12. Juli 1999

CLAIMS

1.) Organic material comprising tris(8-quinolinolato)aluminum(III) (Alq_3) as a base unit,

characterized in that

the base unit is substituted in the 3- or 4- position with an electron-donor group and, simultaneously, in the 5-position with an electron-acceptor or p-delocalizing group.

2.) Material according to claim 1, wherein said electron-donor groups in 3 or 4-position are selected from the group consisting of -CR'R''R''', NR_2, and -OR, whereby R, R', R'' = H or Alkyl, and R'''= Alkyl.

3.) Material according to claim 1 or 2, wherein said electron-acceptor or p-delocalizing groups in the 5 - position are selected from the group consisting of $-CX_3$, $-CX_2-CX_3$, $-SO_3R$, $-CR=CR_2$, $-CX=CX_2$, -COOR, $-SO_3R$, $-SO_3M$ and -COOM, whereby X = F, Cl, Br; R = H or Alkyl, and M = metal ion.

4.) Material according to claim 1, wherein the electron-donor group in the 3- or 4 position is $-CH_3$ and the electron-acceptor group in 5-position is $-CF_3$.

5.) Material according to claim 1, wherein the electron-donor group in 3- or 4-position is -OR and the electron-acceptor group in 5-position is $-CF=CF_2$.

6.) Material according to claim 1, wherein the electron-donor group in 3 or 4 position is -CH_3 and the electron-acceptor-group in 5-position is $-CF=CF_2$.

7.) Use of the material according to any one of the preceding claims in a light emitting device.

8.) An organic electroluminescent device comprising an anode, an organic hole injecting and transporting zone, an organic electron injecting and transporting zone, and a cathode,

characterized in that

said device comprises a luminescent layer comprised of tris(8-quinolinolato)aluminum(III) (Alq_3) , whereby said Alq_3 is being substituted in the 3- or 4- position with an electron-donor group and, simultaneously, in the 5-position with an electron-acceptor or a p-delocalizing group.

9.) Device according to claim 8, wherein said electron-donor groups in 3- or 4-position are selected from the group consisting of -CR'R''R''', NR_2, and -OR, whereby R, R', R'' = H or Alkyl, and R'''= Alkyl.

10.) Device according to claim 8 or 9, wherein said electron-donor or p-delocalizing groups in 5-position are selected from the group consisting of $-CX_3$, $-CX_2-CX_3$, $-SO_3R$, $-CR=CR_2$, $-CX=CX_2$, -COOR, $-SO_3M$ and -COOM, whereby X = F, Cl , Br; R = H or Alkyl, and M = metal ion.

SZ 9-99-004

EPO - Munich
52
12 Juli 1999

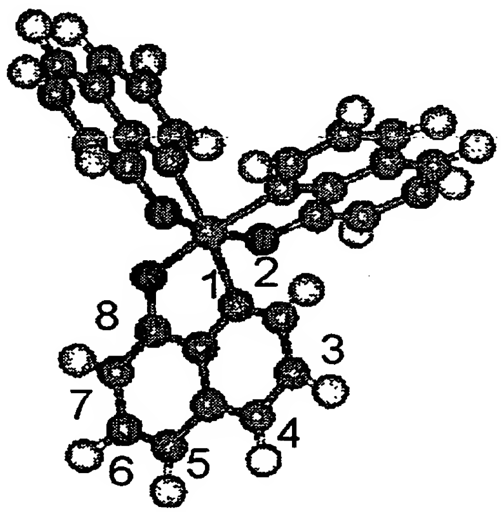

FIG. 1

THIS PAGE BLANK (USPTO)

SZ 9-99-004

EPO - Munich
52
12. Juli 1999

ABSTRACT

An organic material is provided that can be used for a light emitting device. The base unit of said material is tris(8-quinolinolato)aluminum(III) (Alq_3). This Alq_3 is subtituted in the 3- or 4- position with an electron-donor group and simultaneously in the 5-position with an electron-acceptor or p-delocalizing group. Using this material as an emitting luminescent layer, the efficiency of the intrinsic luminescense can be greatly enhanced.

THIS PAGE BLANK (USPTO)